Ultrafast electron thermalization in a magnetic layered Au/Co/Au film

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The ultrafast electron dynamics subsequent to a hundred femtosecond photo-excitation is monitored in a few nanometer thick Au/Co/Au film by means of time-resolved transient spectroscopy. We show that the insertion of the thin Co layer in the Au film leads to a drastic modification of the scattering rates of the different thermalization regimes. The electron-electron interactions are dominated by the mobile *sp*-electrons scattering on the heavy *d*-electrons, which concomitantly enhances the electron-phonon interactions by up to a factor of 5. In the framework of ultrafast magnetization dynamics, this study provides important data on the electronic processes underlying the spin dynamics in a sample typically designed for magneto-optical applications.

PACS numbers:

I. INTRODUCTION

In recent years a number of studies in multilayered nanostructured magnetic devices have paved the way to a wealth of applications. As an outstanding example in the context of information storage technology, most of today's hard drive reading heads are based on the Giant Magneto-Resistance effect (GMR) which was initially observed in a sample made of an alternation of metallic and magnetic nanoscale layers [1]. From a fundamental viewpoint these structures have also allowed important findings on the working of magnetism in a regime comprized between atomic and macroscopic scales.

In connection to information storage technology and to the reading and writing speed limiting processes, a research field has emerged in the last decade which is devoted to ultrafast spin dynamics, namely the possibility of changing and reading the magnetization state of a sample on a sub-picosecond time scale [2–6]. Such a femtosecond demagnetization is much faster than any other process observed so far and in particular much faster than the usual process involving the application of an external magnetic field to drive the magnetization. Although the ultrafast demagnetization effect has been confirmed by several teams using various techniques, some issues related to the microscopic mechanisms involved are still controversial. However it appears that the key role is played by the intricate interactions between the hot electrons, the electron bath and the phonons at a sub-picosecond time scale. Because of their relevance in a number of issues and applications it is important to delineate the underlying processes and assess their impact.

The electron dynamics can be accessed by means of ultrashort light pulse techniques where the system is brought off equilibrium by a sudden photo-excitation and the ensuing evolution of the electron population is monitored by a delayed probe pulse. In the case of noble metals, earlier studies have allowed to map out the athermal regime and the onset of the thermalization below a few hundreds of femtoseconds and beyond respectively [7–11]. An important finding in these single type *sp*-electron systems is the retardation of the intra-electronic thermalization because of the Pauli exclusion principle in a nearly degenerate Fermi liquid and its entanglement with the electron-phonon processes [10–14].

Here we address this issue of the ultrafast electron dynamics in a nanostructured layered sample -such as those of interest for information technology- made of an alternation of Co and Au layers. We show that although the linear optical properties of the sample are almost identical to the ones of pure gold, its electron dynamics is dramatically affected by the presence of the thin Co layer. Both the electron-electron and electron-phonon interactions are enhanced by a factor of five which we interpret in the light of the peculiar electronic properties of transition metals and the specific characteristics of 3*d*-electrons. We measure the average electron thermalization times as a function of the Co layer thickness and show that no interface effect is observed and that the dynamics can be modeled as if the *sp*-electrons were completely delocalized over the whole structure and interacting with 3*d*-electrons acting as scattering centers in many respects similar to external impurities.

II. SAMPLE

The sample consists of a 28 nm thick layer of c.f.c. gold grown in the (111) direction covered by a layer of (0001) h.c. cobalt and capped by 15 atomic monolayers (ML) of gold [15]. The cobalt layer has a step-like structure, each step being 1.6 mm wide and the thickness of the cobalt layer increasing by one atomic monolayer for each step. The thickness of the cobalt layer spreads from 4 up to 12

atomic monolayers. We also use a film of gold on a glass substrate, the thickness of the former being equivalent to the total thickness of the layered sample (31 ±1 nm).

III. EXPERIMENTAL

The pump-probe setup is based on an amplified Ti:Sapphire laser delivering pulses at a central wavelength of 800 nm with a duration of about 250 fs and a repetition rate of 250 kHz. Part of the beam is used as a pump whereas the other part is sent to an homemade Optical Parametric Amplifier which is tunable from 500 nm through 700 nm and serves as a probe. The overall temporal resolution of the setup is about 350 fs (cf. Fig. 2). The focused spots have a diameter of 40 μm for the probe and 50 μm for the pump, ensuring that we probe an almost homogeneously excited region. We record simultaneously the changes of transmission and reflexion of the sample as a function of the time delay using standard synchronous detection methods involving mechanical chopping of the pump beam and lock-in amplification. Part of the probe beam is picked up before the sample and serves as a reference in order to counterbalance the laser intensity fluctuations leading to a highly reduced noise level. This allows us to achieve detection of relative changes of transmission ($\Delta T/T$) and reflexion ($\Delta R/R$) as low as 10^{-4}.

As can be inferred from the skin depths, 15 nm in gold and 5 nm in cobalt at a wavelength of 800 nm, and the values of ϵ''_{Au} and ϵ''_{Co} [16, 17], the same amount of energy is homogeneously deposited in the Co and Au layers but with appropriately different densities. With prevailing electron mean free paths, 30 nm in gold and 5 nm in cobalt, i.e. much larger than the thickness of the cobalt layer, and Fermi velocities in the range of 10^6m.s^{-1}, the excitation can be taken homogeneous over the whole sample thickness after a few tens of femtoseconds with a density of the order of 100 J/cm^{-3}.

IV. MODELING

For the analysis of the experimental results the usual application of electromagnetic wave propagation in stratified dielectric media cannot be transposed as such in the present case [18]. There, the dielectric response being local, the optical coefficients of the different strata are immune to the interfacing. In contrast, in the present case of a metallic structure, the electron are highly delocalized. Taking into consideration the electron characteristics in these two metals (same elementary unit cell volume and *sp*-electron densities), we adopt an embedment approach of the magnetic metal layer into the nonmagnetic ones and assume that all metal layers possess indistinguishable *sp*-electrons uniformly extending over the whole composite film with an intercalated layer of *d*-electrons with markedly different characteristics and that essentially act as scattering centers. Since the *sp*-electron delocalization length is much larger than the layer thickness, we assume an effective Drude-like dielectric contribution for the composite medium. In addition to the above free-electron (intraband) Drude-like contribution, there is a weaker bound-electron (interband) contribution. Practically we take the thickness averaged dielectric contributions from the different layers to account for the effective dielectric function of the sample. It almost reduces to that of pure gold but for the scattering rate which is significantly enhanced by the Co *d*-electron contribution.

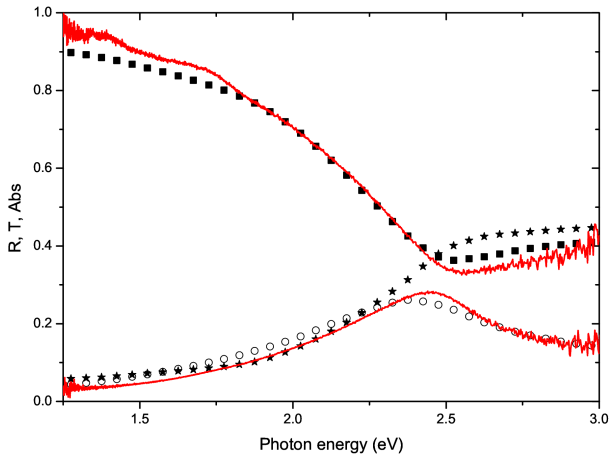

FIG. 1: Transmission (open circles), reflection (black squares) and absorption (stars) of a layered structure consisting of a 28 nm thick gold buffer layer, 6 monolayers of cobalt and 15 monolayers of gold on a glass substrate, computed from the effective medium model using dielectric constants from Ref. [16, 17, 19]. Light scattering has been neglected. Full red line : experimental reflection and transmission coefficients normalized to the values computed at 2.35 eV

We performed a simulation of the optical properties of a composite layered Au/Co/Au film in the framework of the effective medium dielectric response described above, taking as an input tabulated values of the dielectric matrix elements for gold and cobalt [16, 17, 19]. The reflection and transmission coefficients of a structure corresponding to 6 monolayers of Co embedded in a 29 nm thick layer of Au is displayed in Fig. 1 as a function of the photon energy. For low photon energy the layered structure shows the trends of a perfect metal with high reflection coefficient and low absorption. In the visible range the transmission shows a maximum around 2.4 eV which is known as the threshold of the interband transitions in gold. As expected for a sample containing 97% of gold atoms, the overall linear optical properties of the composite material are very similar to those of pure gold. Experimental data points for the reflection and transmission coefficient are displayed in the same figure. The overall agreement with the model is very good. As will be reported elsewhere, this formalism also provides a good estimate of the magneto-optical rotations (Kerr and

Faraday effects), provided that one introduces a nondiagonal dielectric element proportional to the Cobalt one and to the Cobalt layer thickness.

This model (Fig. 1) also allows us to compute the changes of transmission and reflection induced by small changes in the dielectric function of either materials and in particular those resulting from photo-induced changes of the electron population. We find that to an excellent approximation, changes in the dielectric function of cobalt lead to negligible effects as compared to the ones due to changes in the dielectric tensor of gold. Roughly, the relative change of transmission induced by a given change of the dielectric function of gold is 100 times larger than the one induced by the same change in the dielectric function of cobalt. The main conclusion is that the optical properties of the structure basically reflect those of gold, that is those involving the delocalized *sp*-electrons. This corresponds to the fact that the photo-excitation leads to significant changes in the occupation number of the only states lying close to the Fermi level.

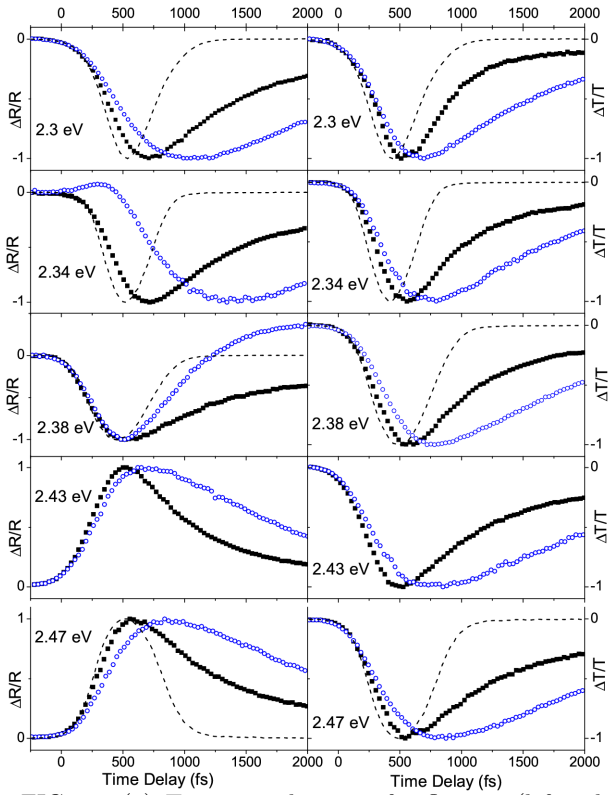

FIG. 2: (a) Transient changes of reflection (left side) and transmission (right side) of a layered Au/Co/Au film (black squares) and of a 30 nm thick reference Au film (open blue circles) for several probe energies. The pump photon energy is kept at 1.55 eV. The dashed line is the pump-probe cross-correlation.

V. EXPERIMENTAL RESULTS

Figure 2 displays the transmission and reflection transients of the layered sample together with those of the reference gold film at different probe wavelengths. Compared to the pump-probe cross-correlation, the transmission transients show a finite rise time (related to the internal thermalization through electron-electron collisions) of the order of one hundred femtoseconds and a recovery time of the order of one picosecond (related to the external thermalization through electron-phonon interactions).

At any probe wavelength we observe that both the rise and decay times are smaller in the layered system than in pure gold. This means that both the electron-electron and electron-phonon interactions are significantly enhanced by the presence of the thin cobalt layer.

In contrast to the transmission transients, the changes of reflection show a strong wavelength dependence in the spectral range investigated in this paper. The reflection transients are negative at low probe photon energy and become positive at high photon energy. For the intermediate photon energies 2.34 eV and 2.38 eV, the gold signal shows a bipolar temporal profile. The results obtained for the gold film are very similar to those reported by Sun *et al.* especially regarding the sign of $\Delta R/R$ as a function of the probe wavelength [8]. This behavior has been accounted for with a two-population model describing both the thermalized and off-equilibrium electron populations. It has been shown that in this probe photon energy range, slightly below the interband transition threshold, the thermalized and out-of-equilibrium populations give contributions to $\Delta R/R$ with opposite signs, which explains the bipolar profiles observed for probe energies of 2.34 eV and 2.38 eV for instance. At lower energies (2.3 eV and below) and at higher energy (2.43 eV and above) $\Delta R/R$ is dominated by the thermalized contribution. Extending this analysis to the case of the layered system gives insight into the electron interactions in this system and confirms the modeling of the effective dielectric response as previously outlined.

VI. INTERNAL THERMALIZATION

A. Results

First, the overall spectral evolution of the transients (sign of $\Delta R/R$) is very similar to the one of the gold film (negative change of reflection at low energy, positive at high energy). This confirms our anticipation that the optical response of cobalt is not distinguishable from that of gold to the extent that it stems from the mobile *sp*-electrons that share the same characteristics. However transients measured in the layered system and in the gold film are qualitatively different in the spectral region where both the thermalized and out-of-equilibrium populations give comparable contributions. Especially

the bipolar profiles observed in $\Delta R/R$ for the gold film at 2.34 eV and 2.38 eV are absent in the layered system. For instance for a gold film probed at 2.34 eV the positive bump (see Fig. 2) has been shown to be due to the off-equilibrium electronic population whereas the negative one is due to the thermalized population [8]. In the layered system, only the negative one remains, i.e. no nonthermal electronic population is observable within our temporal resolution. In fact, because of the presence of the *d*-electrons in the intercalated Co-layer the $sp-d$ collisions are dominant, suppressing the nonthermal electronic population within our temporal resolution. Thus we conclude that the internal electronic thermalization is achevied within 300 fs which corresponds to a strong enhancement of the electron-electron scattering with respect to the case of pure gold. Consistently, at 2.43 eV $\Delta R/R$ becomes positive, as expected from the gold case and still no evidence of any nonthermal population is to be seen.

In both spectral regions where the thermalized contribution is expected to dominate we find that $\Delta R/R$ and $\Delta T/T$ have the same sign as in pure gold. Thus we can analyze the transients for these wavelengths in the light of a two-temperature model which in principle allows to extract both electron-electron and electron-phonon relaxation times [20]. We fit the transients to the convolution of the experimental pump-probe cross-correlation with eq.(1) :

$$X(t) = H(t)\{A(1-e^{\frac{-t}{\tau_{ee}}})e^{\frac{-t}{\tau_{eph}}} + B(1-e^{\frac{-t}{\tau_{eph}}})\} \qquad (1)$$

where $H(t)$ stands for the Heaviside function, τ_{ee} stands for the average *sp*-*d*-electron relaxation time, τ_{eph} stands for the average electron/phonon relaxation time and the second term accounts for the long living plateau corresponding to an overall heating of the sample after electrons and phonons have reached thermal equilibrium. The decay time of this plateau is of the order of several nanoseconds and is not observed on the time scale shown here.

Far off the threshold of interband transitions, we find identical time constants for both the transmission and reflection transients as expected from the two-temperature model. In the case of pure gold at a probe photon energy of 2.47 eV we get $\tau_{ee} = 490$ fs and $\tau_{eph} = 1.1$ ps. These values are in agreement with the ones reported so far in the literature for the low perturbation regime [8, 10]. We checked carefully that these values do not depend on the pump power over one order of magnitude. In the layered system, the rise time of the signal (internal relaxation) is found to be less than 100 fs, roughly 5 times smaller than in bulk gold. However the pump-probe cross-correlation being of the order of 350 fs, our capability to quantify this rise time is limited and hampers any tentative measurement as a function of the Co layer thickness. On a logarithmic scale (see Fig. 3) it is clear that in the layered system the decay becomes exponential earlier than in pure gold, which is consistent with the above mentioned enhanced internal thermalization.

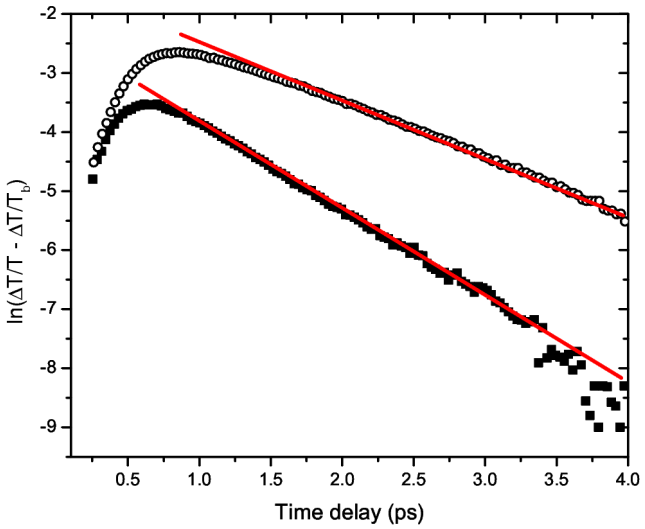

FIG. 3: Logarithm of the transient change of transmission of a layered Au/Co/Au film (black squares) and a reference gold film (open circles). The thermal background has been subtracted. The solid lines are monoexponential fits to the data.

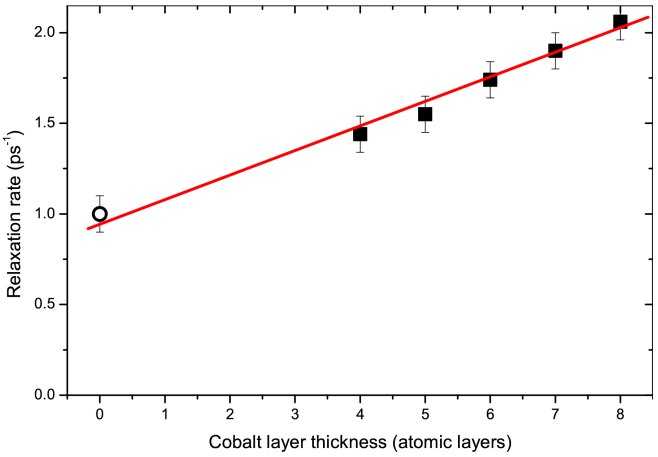

FIG. 4: External (electron-phonon) scattering rate as a function of the cobalt layer thickness (black squares). Scattering rate in the reference gold film (open circle). The solid line is a linear fit to the data.

B. Interpretation

In the electron dynamics, the key role is played by the electron states around the Fermi level and is essentially determined by the broad half-filled *sp*-band which is roughly the same for the two metals. The main difference between noble metals and magnetic ones is in the position of the *d*-bands with respect to the Fermi

level [21–23]. These are much narrower than the *sp*-bands and lie well inside them. In noble metals, they are doubly degenerate, filled and located well below the Fermi level, roughly 2.5 eV in the case of Au. In the magnetic case in contrast, the *d*-bands are only partially filled with the Fermi level cutting through at different positions for spin up and spin down states as the *d*-bands are Zeeman split by the internal magnetization mean field. The *d*-bands being much narrower and less dispersive than the *sp* ones, their density of states is larger by almost a factor five. Thus we may assume the *d*-electrons immobile and not contributing to the dielectric response other than as scattering centers -in several respects similar to static impurities- for the lighter mobile *sp*-electrons. This analysis is a transposition of the two-electron liquid model introduced by Mott for the static/low frequency conductance in magnetic metals to the high frequency/optical dielectric response of these metals [24–26]. The *d*-scattering channels are very efficient and are responsible for the much lower conductivity of magnetic metals as compared to the one of noble metals. From the Fermi golden rule, one can qualitatively argue that the scattering rate of an *s* electron onto *s* and *d* electrons is proportional to their density of states. In the case of Co the latter is about 5 times larger than the former. Although located in the narrow Co layer in our case, these *d*-electrons are efficient scatterers for the *sp*-electrons of the adjacent Au layers as well to the extent that the *sp*-electron delocalization length in Au is larger than the layer thickness, leading to a sizeable increase of the overall scattering rate in agreement with the experimental data.

VII. ELECTRON-LATTICE THERMALIZATION

Similarly, the electron-phonon scattering rate can be deduced from a fit of the data to eq. (1). Since the external relaxation is much slower than the internal one, one can also get an estimate of the electron-phonon scattering rate from a monoexponential fitting of the data for time delays larger than 3 τ_{ee}, after substraction of a long-lasting background due to the final heating of both electrons and lattice (cf. Fig. 3). External relaxation is found to be significantly faster in the layered film than in gold, the relaxation rate depending on the cobalt layer thickness.

Fig. 4 shows the variations of the relaxation rate as a function of the cobalt layer thickness, as well as a linear fit to the data. The extrapolated value of the relaxation rate for a vanishing cobalt layer thickness is very close to the one measured in the reference gold film. This means that interface effects are negligible in the overall relaxation processes in the layered system with respect to bulk effects. This is consistent with our assumption that hot electrons are delocalized over the whole structure.

We can deduce an estimate of the effective electron-phonon scattering rate in the cobalt layer from the variations of the relaxation rate as a function of the cobalt layer thickness. We simply assume that the electron-phonon relaxation rate of the layered structure is given by the average of the relaxation rates in both materials weighted by the probability of presence in each layer. Since the electrons are almost ballistic, this probability is proportional to the thickness of the layer divided by the Fermi velocity in the related material. We use a Fermi velocity of $0.33\text{x}10^6$ m.s^{-1} in cobalt and $1.4\text{x}10^6$ m.s^{-1} in gold. It turns out that the electron-phonon scattering rate in cobalt is of the order of 5 ± 1 ps^{-1} which corresponds to a scattering time of the order of 200 ± 50 fs.

This scattering rate turns out to be about 5 times larger than in noble metals, which is again a direct consequence of the interplay between *sp* and *d* electrons : *d*-electrons being heavy and bound, collisions between *sp* and *d* electrons provide an important path to electron-phonon coupling as discussed above [27]. Data are available in the literature for nickel which is another ferromagnetic transition metal. Although relatively dispersed the values reported so far are in the range of 300 to 800 fs [4, 6, 28, 29]. Cobalt shows a slightly faster electron-phonon scattering time which might be related to its specific band structure around the Fermi level.

VIII. CONCLUSION

In conclusion we have studied the electron dynamics in a layered Au/Co/Au film. We have shown that in the limit of a small cobalt layer thickness, the linear optical properties of the system are almost identical to those of a pure gold film. In contrast, the nonlinear properties are strongly affected by the presence of the transition metal layer. Especially both internal and external scattering rates show a sizeable increase for less than 3 % of cobalt atoms. From a quantitative analysis as a function of the layer thickness, we find that interface processes are negligible. Therefore the accelerated relaxation observed in this structure is due to an intrinsic enhancement of the electronic interactions. This effect is attributed to the large density of *d*-states available near the Fermi level in transition metals. These highly enhanced electronic interactions and the distinct dynamics of the two electron populations represent a valuable input for forthcoming studies about ultra-fast photo-induced spin dynamics in such nanostructured devices.

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